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A second goal was to attempt to clarify a confusing situation in this heterostructure system in which previous photoluminescence measurement[1-3] gave huge discrepancies with predicted emission photon energies of between 50 and 200 meV. This contrasted sharply with the GaAs/GaAlAs system in which agreement was in 1 to 2 meV range[4].

Measurement of Optical Properties of Infrared Bandgap Superlattices

FINAL REPORT

D. F. Nelson

SEPTEMBER 17, 1992

U. S. ARMY RESEARCH OFFICE

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STATEMENT OF PROBLEM STUDIED

The lattice-matched bandgap-tunable heterostructure system $Ga_xIn_{1-x}As_yP_{1-y}/InP$ is technologically important because the bandgap can be tuned with composition between 1.55 and 0.90 μm wavelengths. This range includes two important wavelengths, 1.55 μm where silica glass fibers have their minimum loss and 1.32 μm where such fibers have their minimum group velocity dispersion. These two wavelengths correspond to composition fractions x = 0.47, y = 1 and x = 0.30, y = 0.36 respectively. (The relation x = 0.453y/(1- 0.31 1y) guarantees lattice constant matching of well and barrier.) Devices made with these compositions can be used as laser diodes and photodetector diodes. Thus among the huge number of heterostructure systems presently made, this one deserves special attention.

Because the range of bandgaps in this heterostructure system is further into the infrared when compared to the much studied $GaAs/Ga_xAl_{1-x}As$ system, many spectroscopic techniques are more difficult. For example, prior to this study no inelastic light scattering had been done in this heterostructure system for laser wavelengths longer than the InP bandgap wavelength, that is, in a region where resonant interaction with the confined states of the wells is possible and where four polarization combinations of incident and scattered light are possible (rather than only two when backscattering is done at an opaque wavelength). Utilizing this technique was one of the primary goals of this project.

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erostructure system in which previous photoluminescence measurement[1-3] gave huge discrepancies with predicted emission photon energies of between 50 and 200 meV. This contrasted sharply with the GaAs/GaAlAs system in which agreement was in 1 to 2 meV range[4].

SUMMARY OF MOST IMPORTANT RESULTS

The first stage of this study centered on apparatus development. Automating an inelastic light scattering set-up was the first step. This involved interfacing a SPEX 114 double monochromator and a Thorn-EMI C-10 photon counter (fed by a cooled Hamamatsu R632-01 S-1 photomultiplier) to an AT & T 6300 computer and developing the computer analysis and plotting of data. The second step was developing a set-up to do infrared absorption measurements on the multiquantum well (MQW) samples of InGaAsP/InP. It had been decided that direct absorption measurements would be easier than excitation spectroscopy measurements but would give equivalent data. This is made possible by the fact that MQW samples of InGaAsP/InP are grown on InP substrates, the barrier material. This situation contrasts with the GaAs/GaAlAs system which is grown on GaAs substrates, the well material.

The initial samples studied were grown by molecular beam epitaxy (MBE) by M. Panish of AT & T Bell Laboratories. Because of an extended shutdown of his growth apparatus for relocation to a more safe, isolated building, the next and all succeeding samples were obtained from R. A. Logan of AT & T Bell Laboratories who used the metal-organic chemical vapor deposition (MOCVD) method of growth.

The third preparatory step was the improvement of programs to predict and analyze miniband edges of electrons and light, heavy, and split-off holes in MQWs of varying compositions. These improvements over pro-

grams previously developed by the principal investigator were based on the transfer matrix method (TMM) of L. R. Ram-Mohan of WPI. Ram-Mohan's theory treats four bands (times two for spin), conduction band and heavy, light, and spin- orbit split-off valence bands, in the $\mathbf{k} \cdot \mathbf{p}$ approximation. It accounts for band mixing between the well and barrier materials. Programming the TMM for a superlattice (SL) turned out to be easier than for a single quantum well. A different numerical approach led to calculational stability for single quantum wells and multi-quantum wells (MQW), the latter being the most realistic characterization of the experimental samples. The program was then expanded to calculate not just miniband edge energies but also wavefunctions, well and barrier occupancy fractions, and electron-hole overlap which is a measure of radiative strength. These characteristics help to identify observed transitions.

Our programmed TMM gave us the tool to answer a criticism[5] in the literature concerning previous work of the principal investigator on the effect of bulk band nonparabolicity on SL or MQW energy states. The principal investigator's previous work on this subject as applied to $GaAs/Ga_xAl_{1-x}As(x = 0.39)$ gave both theoretical[6] and experimental[4] support to a modification of Bastard's $\mathbf{k} \cdot \mathbf{p}$ work[7,8,9] which was thus named the "empirical Bastard model" (EBM). The programmed TMM was compared with the simpler, analytical EBM for the circumstances in which the EBM had been challenged. This comparison showed that the EBM was exceptionally accurate (agreement to better than 0.6 meV) for all conduction

band states and, of course, for heavy hole states for which non parabolicity is negligible. When the same numerical comparison was made for light hole valence band states, it was found that the EBM was by far the most accurate analytical model available but nevertheless could be in error by up to 6 meV for the lowest level in a 2 nm wide well or for the highest excited state in a 20 nm wide well. Publication[10] of these TMM calculations thus strongly supported the principal investigator's prior theoretical work[6] and equally strongly refuted the criticism[5] of it.

The improvements that our work with the TMM had led to were also published[11]. The improvements concern the handling of a well-known problem of the $\mathbf{k} \cdot \mathbf{p}$ method, the appearance of spurious solutions with values for the wavevector that lie outside the first Brillouin zone. When using the TMM to calculate a SL configuration in the $\mathbf{k} \cdot \mathbf{p}$ approach, these spurious solutions can be handled quite easily because of the regularity of the SL structure. When a MQW structure or a shaped QW (e.g., parabolic) is considered, however, the spurious solutions must be handled in some way as each successive layer of the structure is calculated by the transfer matrix. This very considerable calculational difficulty was solved by a new analytical technique, the use of a diagonal representation of the transfer matrix. This greatly improved version of the TMM is particularly well suited for $\mathbf{k} \cdot \mathbf{p}$ calculations of arbitrary heterostructures. This allowed testing another controversial aspect in the theory of such structures, the proper boundary conditions between layers of different materials. Some-

what surprisingly, it was found that the range of boundary conditions used in the literature produced only very minor shifts in the energy levels. This helped remove a controversial aspect of this field.

The experimental strategy was to use a tunable laser in the transparent range near the bandgap of InP so that confined states of a MQW could be probed by inelastic light scattering and absorption. Prior to funding under this contract an argon-ion-laser-pumped dye laser was available and initial experiments used this with styryl 9 dye. A commercial Ti:sapphire laser became available at that time and by reprogramming some of the ARO funds, obtaining a grant from the Research Development Council of WPI, and obtaining a discount on the price we were able to purch[illegible] a laser. It has a number of advantages over the styryl 9 dye laser: it emits further into the infrared (1.01 μm wavelength with a 6 W pump), it has no dye degradation, and it operates more stably.

Initial inelastic light scattering measurements on a 30 period MQW sample (120 Å wells and 150 Å barriers) with composition fraction of $x = 0.14$ and $y = 0.30$ showed that the InP Raman scattering dominated the spectrum. To separate InP barrier scattering from InP substrate scattering a right-angle scattering geometry was set up. The incident laser light struck the wafer perpendicular (direction z) to the MQW and near an edge. Magnifying optics imaged the edge of the MQW on the spectrometer slits (scattered light in the y direction). Care was taken to exclude scattered light from the substrate. All four polarization combinations possible in right-

angle scattering were studied: z(xx)y, z(yx)y, z(xz)y, and z(yz)y where the first and last symbols represent the incident and scattered propagation directions and the second and third symbols represent the incident and scattered polarization directions. In the first two cases only Raman scattering from the longitudinal optic (LO) phonons was observed while in the latter two cases only Raman scattering from the transverse optic (TO) phonons was observed. These selection rules differ markedly from those of bulk Raman scattering in a $\bar{4}3m$ binary semiconductor like InP which are z(xx)y, none; z(yx)y, LO + TO; z(xz)y, LO + TO; z(yz)y, TO. The change in selection rules can be understood when it is realized that the InP barrier slabs are thin compared to the wavelength of light, though the calculation is somewhat involved. This was done many years ago by Fuchs and Kliewer[12] and, more relevantly, reconsidered for a MQW recently by Huang and Zhu[13]. Furthermore, these same slab mode selection rules were observed in a GaAs/GaAlAs MQW by Zucker *et al*[14]. There the scattering was from well slabs in contrast to barrier slabs in our work. However, each of the studies observed the phonons of the slab containing the binary semiconductor. This is understandable because Raman scattering from ternary or quaternary alloy phonons is considerably weaker than from binary semiconductor phonons. Our work on the slab mode Raman scattering has been published[15].

We made a determined search for Raman scattering from the quaternary alloy well phonons. They are expected to be very weak because even in a

ternary alloy InGaAs/InP MWQ Watt *et al.*[16] found the Raman scattering from ternary phonons very weak. Nevertheless we were able to observe Raman scattering from a mixed mode[15] at 264 cm $^{-1}$. For the composition of our well material it can be identified from the work of Pinczuk *et al.*[17] on the bulk quaternary material as an unresolved combination of TO and LO modes formed from a mixture of InP and the lattice-matched ternary $In_{0.53}Ga_{0.47}As$ phonons.

Our study of scattering from InP phonons led to observation of very interesting resonance transitions. As the laser photon energy was scanned below the InP bandgap, the strength of the Raman scattering from the InP LO and TO phonons rose sharply as the bandgap energy was approached. This sort of resonance behavior is the same as observed in any bulk semiconductor. Of much greater interest was the observation of two narrow resonances (width 5 meV) at 1.345 and 1.360 eV. Such narrow resonances can only arise from transitions between states of the MQW. However, if the initial and final states are both confined states, the percentage occupancy in the barrier InP material is very low and coupling to the barrier phonons is minimal. Thus we hypothesize that one of the two states is unconfined.

To aid identification of the relevant states we measured the absorption spectrum of the MQW. This revealed transitions at 1.258, 1.264 (not well resolved), 1.298, and 1.311 eV. The rapid rise toward the InP bandgap above these energies made observation of higher confined state transitions uncertain.

Calculations of expected strong ("allowed") transitions using our programmed TMM yielded 1.258 eV ($1e \rightarrow 1hh$), 1.268 eV ($1e \rightarrow 1lh$), 1.292 eV ($1e \rightarrow 3hh$), and 1.317 eV ($2e \rightarrow 2hh$) in good agreement with the absorption transitions. The calculations also revealed two transitions from confined to unconfined states of the MQW at 1.341 eV ($1lh \rightarrow 3e$) and 1.365 eV ($3hh \rightarrow 3e$) that have substantial overlap and hence radiative coupling and substantial barrier material coupling through the unconfined 3e electron miniband. We identify these two transitions with the observed two resonances in the Raman excitation spectrum of InP LO and TO phonons.

The TMM calculations of other resonances at photon energies falling within the Raman excitation spectrum support the interpretation because each of these transitions that possesses significant overlap strength involves two confined states and so has minimal coupling to InP barrier phonons. Thus they are not expected to be observed. These transitions are $3lh \rightarrow 1e$ at 1.348 eV, $2lh \rightarrow 2e$ at 1.351 eV, $5hh \rightarrow 1e$ at 1.355 eV, and $4hh \rightarrow 2e$ at 1.368 eV.

We thus conclude that we have observed Raman scattering resonances of InP barrier LO and TO phonons at transitions between confined and unconfined states of the MQW. We also conclude that our TMM calculation of the $Ga_xIn_{1-x}As_yP_{1-y}/InP$ (x = 0.14, y = 0.30) MQW accounts for the observed transitions to within a few meV. Thus the mysterious 50 to 200 meV discrepancies existing in the literature[1-3] at the time this work began have been eliminated. It should be remarked that the Bell Labs group

found[18] resolution to these discrepancies about the same time that we did. Our work on the resonances of Raman scattering is being prepared for publication[19].

Preparatory to doing laser spectroscopy near the bandgap of the ternary well material $In_{0.53}Ga_{0.47}As$, a tunable color center laser using the F_2^+ center (an ionized double vacancy) stabilized by OH^- dopant in NaCl was constructed and operated by Lixin Wu for his master of science thesis. The laser is pumped by the optical harmonic of a YAG:Nd laser and must be operated with the NaCl crystal held at 77K.

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